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Neutron and X-ray Scattering Group

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Research Activities

Preface

This group has been undergoing a major reconstruction. After leaving of Yasuo ENDOH, who had led the group for many years, to the Institute for Materials Research (IMR) in Tohoku University, we looked for our new group leader. With tremendous support from many faculty members of the physics department, we have succeeded in hiring Youichi MURAKAMI, who had been an associate professor of Photon Factory, KEK, and, more importantly, has been our very important collaborator recently. He has joined the group in April 2001. Until his arrival, Kazuma HIROTA continued to serve as the interim group leader.

These changes have not slowed down our research activities, and will accelerate them. One of our main targets is to construct *Orbital Physics*, and we have already

accomplished various scientific discoveries. In particular, we have utilized a new powerful technique called "resonant x-ray scattering," established by Youichi MURAKAMI in a close collaboration with this group. We are actively working on the orbital degrees of freedom in Mn perovskite oxides as well as the antiferro-quadrupolar ordering in f-electron systems. We have also continued to study Complementary use of neutron and resonant x-ray scattering has clarified important aspects of orbitals in magnetic, structural and transport properties of these systems. Many of these studies have been done through the CREST (Core Research for Evolutional Science and Technology) research group studying the "Dynamical structure of the *orbital* as the novel quantum degrees of freedom," supported by Japan Science and Technology Corp. As a major group of the "High-T_c Consortium" among Tohoku-Kyoto-BNL-MIT, we also have contributed to the research of high temperature superconductivity, particularly the spatial structure of spins and charges in these systems. Reflectivity studies of magnetic thin films and multilayers are also important to us. The polarized neutron reflectometer PORE at KENS spallation neutron source, constructed by Masayasu TAKEDA, has started producing new scientific results in these areas. Studies by PORE also contribute to instrumentation science which is required to design new reflectometers constructed in a coming neutron facility through a joint project by KEK and JAERI.

Scientific activities

Resonant x-ray scattering study of the quadrupolar ordering in RB_2C_2 (R = rare earth)

(T. Matsumura, N. Oumi and K. Hirota)

There are many magnetic materials in which the orbital degrees of freedom play important roles. We have been performing resonant x-ray scattering experiments to observe various behaviour of the quadrupolar moments of the $4f$-electron systems. Following the suceess of the observation of the antiferro quadrupolar ordering in DyB_2C_2, we have extended the study to HoB_2C_2 in which the competition between magnetic and quadrupolar interactions is more important for the physical properties. Temperature dependence of the order paremeter shows that magnetic and quadrupolar orderings of long range appear at the same temperature of 5 K, indicating the crucial role of the quadrupolar moment for the unusual magnetic structure. The work has been done in collaboration with Y. Murakami, H. Nakao (PF, KEK) and Y. Wakabayashi (Keio University).

Vertical Boundary at $x \approx 0.11$ in the Structural Phase Diagram of the $La_{1-x}Sr_xMnO_3$ System ($0.08 \leq x \leq 0.125$)

(K. Hirota, K. Koganei and T. Matsumura)

The structural phase diagram of the $La_{1-x}Sr_xMnO_3$ system in the compositional range $0.08 \leq x \leq 0.125$ has been investigated by high-resolution synchrotron x-ray powder diffraction techniques between 20−600 K. Recent studies have reported that there is an unusual rentrant-type phase transition in this range involving an abrupt

change in lattice parameters but no change in the crystal symmetry, which remains orthorhombic *Pbnm*. The transition to the reentrant phase is from a ferromagnetic metallic to a ferromagnetic insulating phase with some unusual properties. Our results demonstrate that for samples with $x = 0.11 - 0.125$ there exist two lower-symmetry structural regions having monoclinic and triclinic symmetry respectively. There is a sharp first-order transition from the monoclinic to the triclinic phase coinciding with the transition to the ferromagnetic insulating phase, and an abrupt crossover from the orthorhombic *Pbnm* region with a near-vertical phase boundary just below $x = 0.11$. The new phases indicate the presence of some novel type of orbital ordering unlike that found in $LaMnO_3$. We have been also working on the orbital ordering by resonant x-ray scattering and the spin dynamics by inelastic neutron scattering. This work has been done in collaboration with D.E. Cox (BNL), T. Fukuda, J. Mizuki (SPring-8, JAERI), Y. Murakami (PF, KEK) and Y. Endoh (IMR, Tohoku University).

Spin dynamical properties and orbital states of the layered perovskite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ $(0.3 \leq x < 0.5)$

(K. Hirota and K. Shimizu)

Low temperature spin dynamics of the double layered perovskite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (LSMO327) was systematically studied in a wide hole concentration range $(0.3 \leq x < 0.5)$. The spin-wave dispersion, which is almost perfectly two-dimensional, has two branches due to a coupling between layers within a double-layer. Each branch exhibits a characteristic intensity oscillation along the out-of-plane direction. We found that the in-plane spin stiffness constant and the gap between the two branches strongly depend on x. By fitting to calculated dispersion relations and cross sections assuming Heisenberg models, we have obtained the in-plane ($J_{\|}$), *intra*-bilayer ($J_{\perp}$) and *inter*-bilayer (J') exchange interactions at each x. At $x = 0.30$, $J_{\|} = -4$ meV and $J_{\perp} = -5$ meV, namely almost isotropic and ferromagnetic. Upon increasing x, $J_{\perp}$ rapidly approaches zero while $|J_{\|}|$ increases slightly, indicating an enhancement of the planar magnetic anisotropy. At $x = 0.48$, $J_{\|}$ reaches -9 meV, while $J_{\perp}$ turns to $+1$ meV indicating an antiferromagnetic interaction. Such a drastic change of the exchange interactions can be ascribed to the change of the relative stability of the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbital states upon doping. However, a simple linear combination of the two states results in an orbital state with an orthorhombic symmetry, which is inconsistent with the $I4/mmm$ tetragonal symmetry of the crystal structure. We thus propose that an "orbital liquid" state realizes in LSMO327, where the charge distribution symmetry is kept tetragonal around each Mn site. Orbital liquid states are formulated in a theoretical model which takes into account strong electron correlation. The calculated results satisfactorily explain the systematic changes of the exchange interactions in LSMO327 observed in the experiments. This work has been done in collaboration with S. Ishihara, S. Maekawa, Y. Endoh (IMR, Tohoku University), M. Kubota (PF, KEK), H. Yoshizawa (ISSP, University of Tokyo) and Y. Moritomo (CIRSE, Nagoya University).

Neutron-Scattering Study of Zn-doped $La_{2-x}Sr_xCuO_4$

(M. Kofu and K. Hirota)

Neutron-scattering study of static antiferromagnetic correlations has been performed in the overdoped $La_{2-x}Sr_xCuO_4$ (LSCO) with a partial substitution of Zn. In the single crystal of $La_{1.79}Sr_{0.21}Cu_{0.99}Zn_{0.01}O_4$, of which the superconductivity is strongly suppressed ($T_c \leq 2$), incommensurate magnetic peaks were observed as an elastic scattering. It was also confirmed that the elastic peak intensity develops below 20 K, and the incommensurability ε was estimated to be 0.135 ± 0.002 r.l.u., which is much smaller than the hole concentration x (= 0.21). Furthermore, in our preliminary measurements, we observed that the incommensurate peak positions shift away from the [1 0 0] tetragonal axis, which was already observed in underdoped LSCO and excess-oxygen-doped system. We have also grown single crystals near $x \sim 0.15$, which will be studied in the 2001 fiscal year. This work has been done in collaboration with H. Kimura (RISM, Tohoku University), Y. Koike (Applied Physics, Tohoku University), Y. Endoh (IMR, Tohoku University) and K. Yamada (ICR, Kyoto University).

Spin dynamics of NiS_2

(M. Matsuura and K. Hirota)

Neutron scattering experiment on NiS_2 single crystal revealed a honeycomb pattern of the intensity distribution in reciprocal lattice space (continuous-line structure along the *fcc* zone boundary) providing the first direct evidence for nearly frustrated antiferromagnetism (AF) on the face centered cubic (*fcc*) lattice. A small but finite lattice distortion below 30.9 K lifts the degeneracy of the magnetic ground state due to the frustration and eventually result in the coexistence of the type I and the type II AF long-range orderings, which are mutually incompatible in the *fcc* symmetry at higher temperatures. This work has been done in collaboration with H. Hiraka, Y. Endoh (IMR, Tohoku University) and K. Yamada (ICR, Kyoto University).

Relation between conduction electrons and crystalline electric field

(T. Matsumura)

Strong correlation between conduction electrons and a crystalline electric field that localized $4f$ electrons experience was discovered experimentally in $Tm_xLa_{1-x}Te$. In $Tm_{0.05}La_{0.95}Te$ where the Tm ions are diluted in a metallic environment, large crystal field of about 100 K was observed. However, in $Tm_xYb_{1-x}Te$, an insulating system, the crystal field almost vanishs. This difference apparently comes from the existence of the conduction electrons. We study this relation by changing the number of conduction electrons. The experiment was first performed with the HET spectrometer at ISIS, UK, and now being performed at the LAMD spectrometer at KEK with K. Shibata (IMR, Tohoku University).

Magnetic Structure of Thin Film Cr in Cr/Sn Multilayers

(M. Takeda)

Chrominium (Cr) in thin films and multilayers exhibits a variety of magnetic properties different to those of bulk Cr due to low dimensionality, proximity effect, internal strain and so on. We have investigated magnetic structures of Cr/Sn multilayers with monatomic nonmgnetic Sn spacer layer on MgO substrate by neutron scattering using triple-axis spectrometers TOPAN of Tohoku University and TAS-1 of Japanese Atomic Energy Research Institute (JAERI) in JRR-3M in Tokai. Epitaxial Cr/Sn films with different multilayer period, $\Lambda = t_{Cr}+t_{Sn}(t_{Cr} = 4, 8, 12, 16\text{nm}, t_{Sn} = 0.2\text{nm})$ were prepared by ultrahigh-vacuum deposition. The number of stacking was changed so as to have the same total thickness of Cr, 240 nm. Cr in the multilayers has bcc structure and the [001] is perpendicular to the film plane. A commensurate antiferromagnetic (CAF) structure is stabilized at 300 K in these multilayers. With decreasing the temperature the CAF changed into coexistence phase of CAF and spin density wave (SDW) except the sample with $t_{Cr} = 4\text{nm}$. It was found that the SDW period in these multilayer was much longer than that of bulk Cr at the same temperature and were determined by each multilayer period. It has been theoretically predicted that Cr magnetic moments at interface between Cr and non magnetic layer ware enhanced. The enhancement makes antinodes of SDW at the interface. Consequently the SDW whose wave vector is determined not by the nesting vector but by the multilayer period, $\Lambda, 2\Lambda...$, was formed in this multilayer. This work was done in collaboration with K. Mibu and T.Shinjo of Institute for Chemical Research in Kyoto University, J. Suzuki of JAERI, Ki Bong Lee of Pohang University of Science and Technology in Korea and Y. Endoh of Institute for Material Research in Tohoku University.

Specular and Off Specular Neutron Reflection from Magnetic Interfacial Roughness in Multilayers with the GMR effect

(M. Takeda)

Fe/Cr multilayers:

It is well known that a giant magnetoresistance (GMR) effect occurs in magnetic multilayers which are composed of ferromagnetic layers antiferromagnetically coupled across nonmgnetic spacer layers. It has been pointed out that the interfacial roughness is essential to the GMR effect in magnetic multilayers because the spin dependent scattering of conduction electrons at the interfaces are responsible for the GMR effect. The interfacial roughness, atomic disorder at the interfaces, is expected to be accompanied with the magnetic disorder. Such magnetic disorder may directly affect on the GMR effect because the spin-dependent scattering of conduction electrons at the interface is believed to be essential to the GMR effect in this multilayer. However, little attention has been paid to the magnetic disorder at the interface in studying the GMR effect. The correlation between interfacial roughness and the GMR effect in Fe/Cr multilayers were investigated by using neutron reflection, neutron diffraction and X-ray. The specimen were annealed under various condition in vacuum so as to have different interfacial roughness. Clear off-specular reflection

which indicated the existence of correlated interfacial roughness from the bottom to the top layer was observed in these multilayers. The off-specular reflection around the antiferromagnetic peaks was very sensitive to the interfacial roughness, on the other hand, that around the Fe/Cr bilayer peak was little affected by the roughness. This suggests that the magnetic interfacial roughness is more essential to the GMR effect than the atomic interfacial roughness. This work was done in collaboration with A. Kamijo of NEC corp., Ki Bong Lee of Pohang University of Science and Technology in Korea, S. Langledge of ISIS in UK and Y. Endoh of IMR in Tohoku University.

Magnetic Interfacial Roughness in Co/Cu Multilayers Fabricated by Sputtering in Ultra Clean Ar Gas:

In magnetic thin films and multilayers it is known that purity of the sputtering atmosphere strongly affects the magnetic properties associated with the significant change of microstructure. Recently we have fabricated Co/Cu multilayers by the sputtering in the chamber in which the residual impurities were carefully controlled by changing the pressures, P_b, just before introducing processing gas of ultra-clean Ar. It is found out that the magnetoresistance ratio (MR) was drastically dropped from 48 % to 14 % with decreasing slightly from 7×10^{-8} to 3×10^{-8} Torr. This is because the interfacial roughness was enhanced by the purification and weakened the antiferromagnetic coupling energy between the adjacent ferromagnetic Co layers. The decrease of the coupling led to the abrupt drop of the GMR. However, it is worth while to consider the another factor for the abrupt decrease of the GMR in the different point of view; the interfacial roughness accompanied with the magnetic disorder. On a polarized neutron reflectometer, CRISP, in ISIS we have done specular and off-specular reflectivity measurements of two Co/Cu multilayers fabricated by the Ar sputtering under the slight difference of partial pressure of residual oxygen gas, $P_{O_2} = 1 \times 10^{-8}$ and $P_{O_2} = 5 \times 10^{-8}$. The MR ratios, $\rho/\rho(13\ \mathrm{kOe})$, are 14 % and 52 %, respectively. The information of magnetic disorder at interface is give by off-specular reflectivity profile. A correlated interfacial roughness gathers around the Bragg reflection from antiferromagnetic Co/Cu bilayers. We have done the off-specular reflectivity measurements using a 2D PSD, however, we could not see clear off-specular reflections in both samples. The undetectable off-specular reflection suggests that the magnetic interfacial roughness little correlates each other. Even in the case the interfacial roughness can still be estimated by the static Debye-Waller factor for the specular reflection if it exists. The detailed analysis is now in progress. This work was done in collaboration with M. Tsunoda, M. Takahashi of Dept. of Electro. Engineer. and Y. Endoh of IMR in Tohoku University.

Structural and Magnetic Properties of High Saturation Induction CoNiFe Films

(M. Takeda)

CoNiFe electroplated films are promising candidates of write core-material for high-density recording because they have high saturation induction (B_s) and excellent soft magnetic properties. It has been revealed that the magnetic properties are very sensitive to the plating condition, however, factors and mechanism which in-

duce the high B_s are still open questions. We have investigated magnetic structure of these films using neutron reflection and diffraction to reply the questions using a polarized neutron reflectometer, PORE, at KENS and TOPAN at JRR-3M in Tokai, respectively. The films are grown on NiFe buffer layer on Si substrate. Polarized neutron reflectivity gives information on the depth profile of density of magnetic moments, which makes it possible to determine the magnitude of magnetic moment in each layer separately. The films composed of mixed bcc and fcc phases. A band calculation showed that the magnetic moments in bcc phase is slightly larger than that in fcc phase. The neutron diffraction measurements have been done to confirm the difference of magnetic moment in each phases. We are now collecting the data of reflectivity and diffraction pattern from films electroplated under various plating condition. This work was done in collaboration with N. Ohsima, M. Saito, K. Ohashi, A. Kamijo in NEC Corp. and Y. Endoh of IMR in Tohoku University.

Instrumentation

Measurement of Hall effect and magnetoresistivity for studies of magnetism

(T. Matsumura, K. Chatani and T. Muraki)

To study transport properties of magnetic materials as a tool to understand magnetic and orbital correlations, we have developed a measurement system which is able to measure Hall resistivity and magnetoresistivity between 1.5 K and 300 K up to 10 Tesla. Two samples can be measured simultaneously. Temperature control, magnetic field sweep, and sample rotation are computer controled using LabView. $Cr_{1-x}Ru_x$, $La_{2-2x}Sr_{1+2x}MnO_4$, $La_{2-x}Sr_xCuO_4$, and TmX (X=S, Se, Te) are being studied.

Investigation of Magnetism in Magnetic Thin Films and Multilayers by a Polarized Neutron Reflectometer, PORE, at KENS

(M. Takeda)

We are studying magnetic thin films and multilayers using a polarized neutron reflectometer, PORE, at KEK in Tsukuba. This reflectometer is maintained by our group and open to all scientists from the universities and laboratories in the world. In this fiscal year structure of lipid membranes, block copolymers and domain structure of reentrant spin glass were also investigated using this reflectometer. This work was done in collaboration with N. Torikai, T. Otomo, M. Furusaka, S. Satoh of KEK and Y. Endoh of IMR in Tohoku University.

A New Thermal Neutron Reflectometer for Free Surface at KENS – ARISA–

(M. Takeda)

We have installed a new neutron reflectometer, Advanced Reflectometer for Interface and Surface Analysis (ARISA), with vertical scattering-plane geometry for studying free surface at a thermal neutron port viewing an ambient-temperature water moderator at KENS (pulsed neutron source in KEK). ARISA is a unique reflectometer using thermal neutrons at a pulsed spallation neutron source as well as the first neutron reflectometer with vertical scattering-plane geometry in Japan. The designed specifications for the covered range of neutron momentum transfer, Q_z, in the vertical direction are 0.008Å^{-1} - 0.61Å^{-1} for liquid samples and 0.008Å^{-1} - 2.8Å^{-1} for solid ones, respectively. The reflectometer is still in commissioning, however, several experiments have been done and reflectivities down to 10^{-6} have been successfully measured. This work was done in collaboration with N. Torikai, M. Furusaka in KEK, H. Matsuoka, S. Tasaki in Kyoto University, Y. Matsushita in Nagoya University, M. Shibayama of ISSP in University of Tokyo, A. Takahara of IFROC in Kyushu University and H. Yamaoka in University of Shiga Prefecture.

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Master and Doctral Thesis

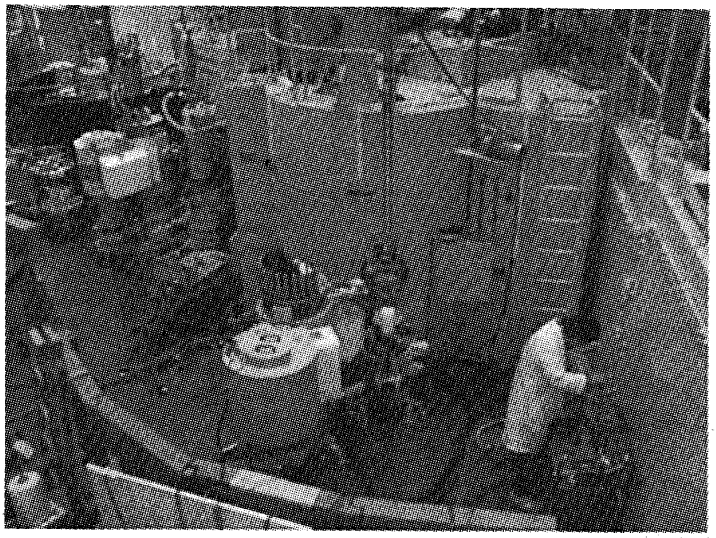
M1) Nobuyuki OUMI: Resonant x-ray scattering study of antiferro quadrupole ordering in RB_2C_2 (R = Dy, Ho).

M2) Yo'uhei KOGANEI: Charge and orbital ordering near the insulator to metal phase boundary in $La_{1-x}Sr_xMnO_3$.

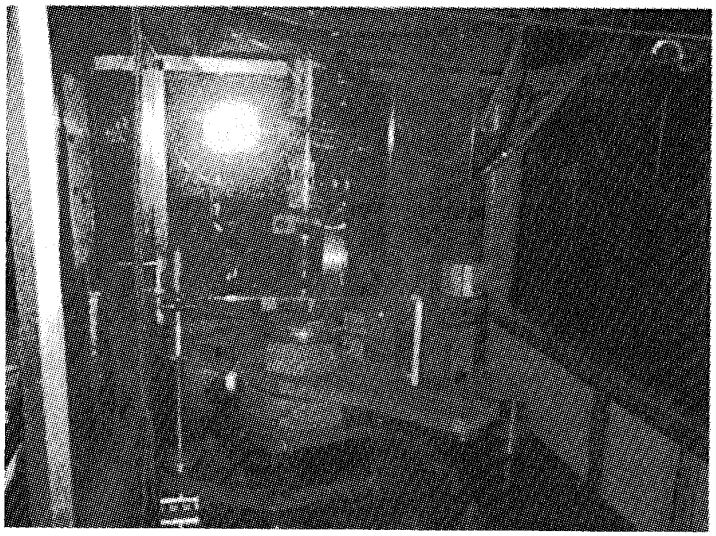
M3) Kouichirou SHIMIZU: Competitive charge/spin orders and phase separtaion in the highly doped region of the bilayer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$.

D1) Kenji KURAHASHI: Large Single Crystal Growth of Electron-Doped Cuprates and Study of Correlation between Superconducting Mechanism and Spin Fluctuations.

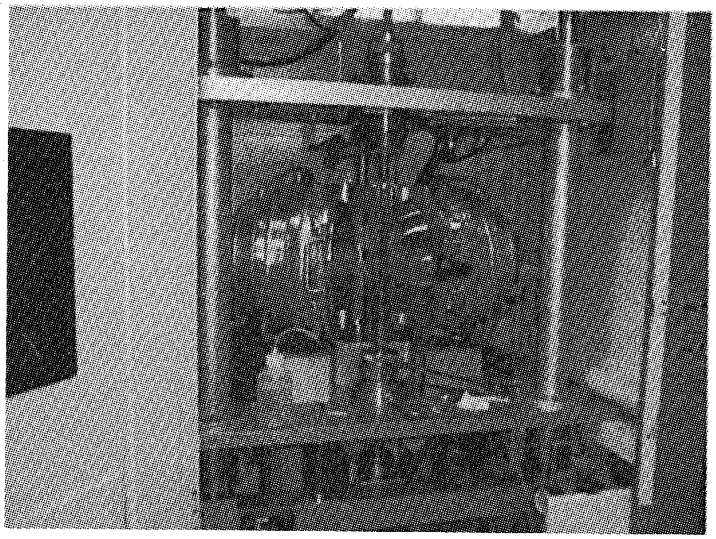
D2) Masato MATSUURA: Magnetic properties of the Mott transition in $NiS_{2-x}Se_x$ – Neutron scattering study of the spin dynamics.



TOPAN (Tohoku University Poralization Analysis Neutron Spectrometer)



Four-circle x-ray diffractometer with ^{4}He closed-cycle cryostat



Floating Zone Furnace